



Organophosphate esters in indoor dust from 12 countries: Concentrations, composition profiles, and human exposure

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ABSTRACT

A total of 20 organophosphate triesters (OPEs), including seven alkyl-OPEs, three chlorinated (Cl)-OPEs, seven aryl-OPEs, and three oligomeric-OPEs were measured in 341 house dust samples collected from 12 countries during the period 2010–2014. OPEs were ubiquitous in indoor dust, and the total concentrations of OPEs (Σ OPEs; sum of 20 OPEs) ranged from 49.4 to 249,000 ng/g dry weight (dw). Generally, Cl-OPEs were the predominant compounds (51% of total) in indoor dust samples, with a median concentration of 800 ng/g, followed by alkyl-OPEs (31%), aryl-OPEs (17%), and oligomeric-OPEs (1%), with median concentrations of 480, 270, and 21.9 ng/g, respectively. Σ OPE concentrations in indoor dust from more industrialized countries (South Korea: median, 31,300; Japan: 29,800; and the United States: 26,500 ng/g dw) were one or two orders of magnitude higher than those from less industrialized countries (Greece: 7140, Saudi Arabia: 5310, Kuwait: 4420, Romania: 4110, Vietnam: 1190, China: 1120, Colombia: 374, India: 276, and Pakistan: 138 ng/g dw). Statistically significant positive correlations ($0.114 < r < 0.748$, $p < 0.05$) were found among the concentrations of 16 OPEs in dust samples, indicating similar sources of these compounds. The median estimated daily intakes of Σ OPEs via dust ingestion for children and adults were in the ranges of 0.29–64.8 and 0.07–14.9 ng/kg bw/day, respectively.

1. Introduction

Organophosphate esters (OPEs) are used extensively worldwide as flame retardants, plasticizers, and anti-foaming agents in various commercial and industrial products, such as electronic equipment, paints, bedding, textiles, furniture, lacquers, floor polishes, and building materials (He et al., 2018). Due to the global restriction and phase-out of polybrominated diphenyl ethers (PBDEs) and

hexabromocyclododecanes (HBCDs), production and consumption of OPEs have sharply increased from 102,000 t in 1992 to 680,000 t in 2015 (Luo et al., 2016; Sun et al., 2018; Zheng et al., 2016). Because most OPEs are physically mixed rather than chemically bound to products, they can emit into the indoor environment from various consumer products via volatilization and abrasion (Cao et al., 2014; Dirtu et al., 2012; Rauert et al., 2014).

Indoor dust is a repository for OPEs released from consumer

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products (Kademoglou et al., 2017). Studies have reported on the concentrations of OPEs in indoor dust (Kademoglou et al., 2017; Shoeib et al., 2019; Wong et al., 2017; Yang et al., 2014), which are considerably higher than those found in sediment (Chen et al., 2018; Zhong et al., 2018), sludge (Gao et al., 2016; Wang et al., 2019), soil (Fries and Mihajlovic, 2011; Yadav et al., 2018) and food (Ding et al., 2018; Zhang et al., 2016). Dust ingestion can be an important OPE exposure pathway in humans. Toxicological studies have shown that some OPEs are neurotoxic, carcinogenic, and endocrine disruptors. For example, tris(2-chloroethyl) phosphate (TCEP), and tris(1-chloro-2-propyl) phosphate (TCIPP) elicited moderate acute toxicity in rats (Lu et al., 2017; Zhang et al., 2018a). TCEP was carcinogenic in both rats and mice (European Union, 2009). In vitro, tris (1,3-dichloro-2-propyl) phosphate (TDCIPP) was identified to possess neurotoxic properties on PC12 cells (Ospina et al., 2018). Tris(2-butoxyethyl) phosphate (TBOEP) caused developmental toxicity in zebrafish (Han et al., 2014). Tri-*n*-butyl phosphate (TNBP) and triphenyl phosphate (TPHP) have been reported to be neurotoxic to zebrafish larvae and rats (Behl et al., 2015; Shi et al., 2018). Thus, OPEs in indoor dust may pose a threat to human health, and studies on human exposure to these chemicals are needed (Khan et al., 2016; Zhang et al., 2018b).

Several studies have reported the occurrence of OPEs in geographically remote regions, such as the Arctic and Antarctic (Möller et al., 2012; Salamonova et al., 2014). Indoor dust is an important source and pathway for OPEs found in the outdoor environment via dispersion (Marklund et al., 2003). Accordingly, indoor dust can play a role in the transportation of OPEs in the environment. A few multinational studies have reported the occurrence of OPEs in indoor dust. Wong et al. (2017) reported the concentrations of 13 OPEs in 24 dust samples collected from five countries (Australia, the United Kingdom [UK], Canada, Sweden, and China), Shoeib et al. (2019) reported the occurrence of 12 OPEs in 148 house dust collected from three countries (Canada, Egypt, and Turkey), and Kademoglou et al. (2017) reported the concentrations of 16 OPEs in 32 indoor dust samples collected from two European countries (the UK and Norway). In this study, we conducted a multinational survey of 20 OPEs, comprising seven alkyl-OPEs, three Cl-OPEs, seven aryl-OPEs, and three oligomeric-OPEs, in 341 indoor dust samples collected from 12 countries (China, Colombia, Greece, India, Japan, Kuwait, Pakistan, Romania, Saudi Arabia, South Korea, the United States [US], and Vietnam). Human exposure to OPEs via dust ingestion by children and adults was estimated based on the measured concentrations. This is the first study to report a broader geographic occurrence (including several Asian countries) and distribution of OPEs in indoor dust from 12 countries.

2. Materials and methods

2.1. Chemicals and reagents

Trimethyl phosphate (TMP), bisphenol A bis(diphenyl phosphate) (BDP), tetrakis(2-chloroethyl)dichloroisopentyl diphosphate (V6), and resorcinol bis(diphenyl phosphate) (RDP) were purchased from Toronto Research Chemicals (North York, ON, Canada). 2-Ethylhexyl diphenyl phosphate (EHDP) and tri-*iso*-butyl phosphate (TIBP) were purchased from Sigma-Aldrich (St. Louis, MO). Triethyl phosphate (TEP), tripropyl phosphate (TPRP), TNBP, TBOEP, tris(2-ethylhexyl) phosphate (TEHP), TCEP, TCIPP, TDCIPP, TPHP, trimethylphenyl phosphate (TMPP), isodecyl diphenyl phosphate (IDDP), cresyl diphenyl phosphate (CDPP), *tert*-butylphenyl diphenyl phosphate (BDPP) and tris(*p*-*tert*-butylphenyl) phosphate (TBPHP) were purchased from AccuStandard (New Haven, CT). Nine isotopically labeled compounds were used as surrogate standards, which comprise, TEP-d15, purchased from Sigma-Aldrich; TMP-d9 and TEHP-d51, purchased from Toronto Research Chemicals; and TPRP-d21, TNBP-d27, TCEP-d12, TCIPP-d18, TDCIPP-d15, and TPHP-d15, purchased from Cambridge Isotope Laboratories (Tewksbury, MA). Detailed information on target compounds is listed in

Table S1 (Supporting Information).

HPLC-grade methanol (99.99%) was purchased from Fisher Scientific (Fair Lawn, NJ). HPLC-grade water, acetonitrile (99.9%), ethyl acetate (99.6%), acetone (99.6%), pesticide grade *n*-hexane (95%), and anhydrous sodium sulfate were purchased from J. T. Baker (Phillipsburg, NJ).

2.2. Sample collection

In total, 341 indoor dust samples were collected from 12 countries, including China ($n = 50$), Colombia ($n = 45$), Greece ($n = 30$), India ($n = 36$), Japan ($n = 14$), Kuwait ($n = 24$), Pakistan ($n = 25$), Romania ($n = 23$), Saudi Arabia ($n = 31$), South Korea ($n = 30$), the US ($n = 15$), and Vietnam ($n = 18$) during the period 2010–2014. The details of the sampling locations are shown in Table S2 (Supporting Information). Sampling was performed by following a common protocol to reduce potential confounding factors. In this study, only house dust samples collected from the bedrooms and living rooms were analyzed. Settled dust samples from the floors in homes were collected by vacuuming for 10 min in the living areas using a household vacuum cleaner or with the use of a brush. After sampling, all dust samples were wrapped in aluminum foil, stored in sealed containers, and stored at -4°C until they were transported to the laboratory. All samples were ground, sieved, and homogenized by passage through a $150\text{-}\mu\text{m}$ sieve, packed in clean aluminum foil, and stored at 4°C in the darkroom until analysis.

2.3. Sample pretreatment

Target compounds were extracted from dust samples and purified by solid-phase extraction with Oasis HLB cartridges (6 mL, 200 mg; Waters, Milford, MA), as described in our previous study, with some modifications (Kim et al., 2019). One hundred milligrams of dust sample were weighed into a 15-mL polypropylene (PP) tube. After spiking with 10 ng of internal standards, dust samples were equilibrated for 30 min at room temperature and extracted twice by shaking in an orbital shaker (Eberbach Corp., Ann Arbor, MI) with 5 mL of methanol for 12 h each time. After shaking, samples were centrifuged at 4500 g for 10 min (Eppendorf Centrifuge 5804, Hamburg, Germany), and the supernatant was transferred into a 15-mL PP tube. The concentrated extracts were purified by passage through Oasis HLB cartridges (6 mL, 200 mg; Waters) that were preconditioned with 5 mL methanol. Cartridges were then eluted with 2 mL of methanol and 1 mL of ethyl acetate. The extracts were concentrated to near dryness and re-dissolved in 0.25 mL of water/methanol mixture (40/60, v/v), and micro-centrifuged (10,000 g, 1 min) ($0.2\text{-}\mu\text{m}$ nylon filter, Spin-X, Costar, Corning Inc., Corning, NY) before injection into HPLC-MS/MS.

2.4. Instrumental analysis

The concentrations of OPEs were determined by an Agilent 1100 HPLC system (Agilent Technologies, Santa Clara, CA) coupled with an API 2000 triple-quadrupole mass spectrometer (MS/MS), equipped with an electrospray ionization source (ESI) (Applied Biosystems, Foster City, CA). An analytical column (Betasil C_{18} , $2.1\text{ mm} \times 100\text{ mm i.d.}$, $5.0\text{ }\mu\text{m}$, Thermo, Waltham, MA), connected to a guard column (Betasil C_{18} , $2.1\text{ mm} \times 20\text{ mm i.d.}$, $5\text{ }\mu\text{m}$; Thermo) was used for LC separation. The mobile phase was methanol with 0.1% acetic acid (A) and HPLC-grade water with 0.1% acetic acid (B) maintained at a flow rate of 0.20 mL/min with a gradient program as follows: 0–2 min, 60% A; 2–7.5 min, 60–99% A; 7.5–17 min, 99% A; 17–17.5 min, 99–60% A; 17.5–25 min, 60% A. The positive ion multiple reaction monitoring mode was used for the identification and quantification of OPEs. Detailed information with regard to tandem mass spectrometry parameters is given in Table S3.

2.5. Quality assurance/quality control

All glassware and PP tubes were rinsed with hexane, acetone, methanol, and acetonitrile to avoid potential contamination during analysis. The internal standard method was used in the quantification of each target compound. An 11-point calibration curve was constructed with OPE standard solutions at a concentration range of 0.10–400 µg/L [0.1, 0.5, 1, 2, 5, 10, 25, 50, 100, 200, and 400 µg/L in water–methanol (4,6, v:v)], and the calibration curves exhibited excellent correlation coefficients ($r^2 > 0.99$). The instrumental limits of detection (LODs) and limits of quantitation (LOQs) were set at a signal-to-noise ratio of 3 and 10, respectively (Table S4). The method detection limits (MDLs) were estimated from LOQs by taking into account a sample amount of 100 mg and a dilution factor of 1:4 (v/v). Procedural blanks were analyzed to check for contamination that may occur during sample preparation and analysis. Trace levels of TNBP, TCIPP, TEHP, TPHP, TMPP, and IDDP (approximately 0.06–0.72 ng) were found in procedural blanks, and background subtraction was performed in the quantification of concentrations in samples. A 10-µg/L OPE standard mixture was injected after every 10 samples to ensure instrumental stability and calibration. Average recoveries of OPEs in spiked blanks and matrices ranged from 74.2 to 116% and from 68.1 to 137%, respectively. Detailed information about recoveries, correlation coefficients, and LODs of target compounds is listed in Tables S4 and S5 in the Supporting Information.

2.6. Statistical analysis

Statistical analyses were performed with SPSS/PASW Statistics 18.0 (SPSS Inc., 1993–2007). Concentrations of target compounds below the MDLs were assigned a value of 1/2 MDLs for data analysis. The total concentrations of seven alkyl-OPEs, three Cl-OPEs, seven aryl-OPEs, three oligomeric-OPEs, and the sum of 20 OPEs were denoted as ΣAlkyl-OPEs, ΣCl-OPEs, ΣAryl-OPEs, ΣOligo-OPEs, and ΣOPEs, respectively. A nonparametric Spearman rank correlation test (two-tailed, significance level at 0.05) was performed to assess the relationships among the concentrations of individual OPEs in indoor dust samples.

3. Results and discussion

3.1. Concentrations

The concentrations and detection frequencies of OPEs in indoor dust from 12 countries are presented in Tables 1 and 2. A total of 20 OPEs were detected in 341 indoor dust samples. Except for TMP, TPRP, TBPHP, RDP, and V6, other OPEs were found at detection frequencies higher than 73%, indicating their widespread usage in the indoor environment of several countries. The concentrations of ΣOPEs, ΣAlkyl-OPEs, ΣCl-OPEs, ΣAryl-OPE, and ΣOligo-OPE in indoor dust samples from 12 countries are shown in Table 2. The total concentration of OPEs (ΣOPEs) in indoor dust ranged from 49.4 to 249,000 ng/g dry weight (dw). Generally, Cl-OPEs were the predominant compounds (accounting for 51% of ΣOPEs) in indoor dust samples, with a median concentration of 800 ng/g, followed by alkyl-OPEs (31%), aryl-OPEs (17%), and oligo-OPEs (1%), with median concentrations of 480, 270, and 21.9 ng/g, respectively.

3.1.1. Cl-OPEs

Due to their high production, extensive usage and persistence in the environment (Guo et al., 2017; Kim et al., 2014; Reemtsma et al., 2008), Cl-OPEs were the dominant OPEs found in indoor dust. Among three Cl-OPEs measured, TCIPP was the most abundant compound, found at a median concentration of 222 ng/g (< MDL-66,700 ng/g), followed by TCEP and TDCIPP at 201 ng/g (< MDL-45,900 ng/g) and 79.6 ng/g (< MDL-16,600 ng/g), respectively. The elevated concentration of TCIPP found in indoor dust was consistent with its high

Table 1

Overall concentrations of organophosphate esters in 341 indoor dust samples collected from 12 countries (ng/g dw).

Compounds	MDL ^a	Min	Median	Mean	95th	Max	df (%) ^b
TMP	0.32	< MDL	< MDL	1.40	1.16	332	11.4
TEP	1.2	< MDL	24.5	116	397	7340	82.7
TPRP	2.1	< MDL	< MDL	3.37	3.38	592	10.0
TNBP	0.95	< MDL	32.5	86.2	373	1690	98.2
TIBP	0.56	< MDL	9.64	37.1	117	1450	98.2
TBOEP	1.87	< MDL	190	7740	50,900	234,000	99.7
TEHP	2.02	< MDL	37.1	84.3	316	2930	93.8
TCEP	0.79	< MDL	201	1260	6880	45,900	99.1
TCIPP	2.33	< MDL	222	1310	5550	66,700	76.2
TDCIPP	0.90	< MDL	79.6	1440	4220	166,000	81.2
TPHP	0.40	< MDL	112	726	2680	25,200	97.4
TMPP	0.35	< MDL	16.5	125	323	7330	98.5
EHDPP	0.29	< MDL	27.3	96.5	396	1920	99.4
CDPP	1.0	< MDL	15.7	316	432	38,900	94.4
IDDP	0.31	< MDL	8.21	171	260	13,600	97.7
BPDP	0.28	< MDL	3.88	17.3	62.5	716	90.9
TBPHP	1.56	< MDL	< MDL	22.3	23.9	5160	22.6
RDP	0.74	< MDL	< MDL	53.7	172	5480	46.9
BDP	0.52	< MDL	3.45	37.7	189	1390	73.3
V6	0.64	< MDL	0.75	124	210	23,500	51.3
ΣAlkyl-OPEs	–	17.0	480	8070	51,300	235,000	100
ΣCl-OPEs	–	2.01	800	4010	16,000	176,000	96.2
ΣAryl-OPEs	–	3.63	270	1470	4400	64,900	99.1
ΣOligo-OPEs	–	< MDL	21.9	215	703	23,600	79.5
ΣOPEs	–	49.4	2220	13,800	79,100	249,000	100

^a Method detection limits.

^b Detection frequency (%).

production volume. TCIPP accounted for about 80% of Cl-OPE usage in Europe and the US (Castro-Jimenez et al., 2014). It should be noted that TCIPP was found in only 76% of the samples, while TCEP, which has gradually been replaced by TCIPP in some regions (due to the carcinogenic and mutagenic properties of TCEP) (Green et al., 2008), was found in 99% of the samples. The relatively high concentrations and detection frequencies of TCEP suggest continuing usage of this chemical in Asia and North America (Ma et al., 2017). In addition, TCEP was found at concentrations of 14% (by weight) in V6 commercial mixtures (Fang et al., 2013). Thus, TCEP found in indoor dust also may originate from the use of V6 in consumer products, although V6 was found only in 51% of the samples. In comparison to the other two Cl-OPEs, TDCIPP was found at relatively low concentrations (median: 79.6 ng/g). This may be ascribed to its high cost, which is about twice that of TCIPP. Another reason is that TDCIPP is used mainly in the automotive industry (outdoor), while only a small fraction is used in furniture (indoor) (Danish Environmental Protection Agency, 2016).

3.1.2. Alkyl-OPEs

All seven alkyl-OPEs were detected in house dust samples (Table 1). TBOEP was the dominant alkyl-OPE, found at a median concentration of 190 ng/g (< MDL-23,4000 ng/g, DF = 99.7%), accounting for 64.4% of the total alkyl-OPE concentrations. This may be related to its widespread use in the indoor environment, including floor polishes, synthetic rubber, plastics, and lacquers (Volkel et al., 2018) (Table S6). TEHP, TNBP, TEP, and TIBP also were frequently detected (DF > 93%) at median concentrations of 37.1, 32.5, 24.5, and 9.64 ng/g, respectively. In contrast, TMP and TPRP were found at low levels with detection frequencies below 12%. TMP is released primarily from industrial processes and seldom found in the indoor environment (Marklund, 2005). TPRP is commonly used as a flame retardant in combination with PBDEs and other OPEs (Van der Veen and de Boer, 2012).

3.1.3. Aryl-OPEs

All seven aryl-OPEs were detected in house dust samples. TPHP was

Table 2
Concentrations of organophosphate esters (OPEs) in indoor dust collected from 12 countries (ng/g dw).

Groups	Compounds	China (50 ^a)	Colombia (45)	Greece (30)	India (36)	Japan (14)	Korea (30)	Kuwait (24)	Pakistan (25)	Romania (23)	Saudi Arabia (31)	USA (15)	Vietnam (18)
Alkyl-OPEs	TMP	Median/df ^b range < MDL-16 < MDL-4.12	< MDL-4 < MDL-1.34	< MDL/0 < MDL	< MDL/25 < MDL-0.57	< MDL/14 < MDL-1.93	< MDL/17 < MDL-2.67	< MDL/8 < MDL-36.7	< MDL/16 < MDL-2.76	< MDL/0 < MDL	< MDL/16 < MDL-7.77	< MDL/13 < MDL-5.52	< MDL/11 < MDL-332
	TEP	Median/df Range < MDL-236 < MDL-10	17.0/78 < MDL-444 < MDL-236	39.6/93 < MDL-2520 < MDL/7	5.69/53 < MDL-3280 < MDL/3	34.9/93 < MDL-902 < MDL/21	141/97 < MDL-659 < MDL/10	18.2/88 < MDL-695 < MDL/21	< MDL/20 < MDL-24.5	44.1/100 3.08-575	26.2/100 5.59-406	74.1/100 8.14-482	37.0/100 1.89-7340
	TPRP	Median/df Range < MDL-24.5 < MDL-10	< MDL-9.55 < MDL-236	< MDL/7 < MDL-27.7	< MDL-2.76 < MDL-3	< MDL-592 < MDL/21	< MDL-9.01 < MDL/10	< MDL-26.9 < MDL/21	< MDL/0 < MDL	< MDL/0 < MDL	< MDL/16 < MDL-3.41	< MDL/13 < MDL-21.0	< MDL/0 < MDL
	TNBP	Median/df Range < MDL-22.100 < MDL-10	4.82/100 < MDL-9.55	66.6/100 < MDL-27.7	7.39/100 < MDL-3	78.8/100 < MDL-592	130/90 < MDL-1690	33.6/92 < MDL-1050	6.56/100 < MDL-28.7	84.0/100 44.4-963	42.3/97 < MDL-156	89.2/100 32.5-1320	16.6/100 2.60-12.6
	TIBP	Median/df Range < MDL-22.100 < MDL-10	1.71-306 < MDL-9.55	1.67/100 < MDL-27.7	1.23-144 < MDL-3	13.0-507 < MDL-592	28.0/90 < MDL-1690	20.1/92 < MDL-1050	5.62/100 < MDL-28.7	47.1/100 21.3-1130	14.7/100 2.61-49.1	13.6/100 1.23-238	3.83/98 < MDL-16.8
	TBOEP	Median/df Range < MDL-22.100 < MDL-10	0.33-8.01 < MDL-9.55	6.17-1060 < MDL-27.7	0.42-32.5 < MDL-3	1.62-45.4 < MDL-592	< MDL-756 < MDL-1450	< MDL-1450 < MDL-1450	0.29-43.5 < MDL-28.7	71.2/100 99.3-19,200	629/100 21.6-4400	12,200/100 575-51,100	103/100 52.3-197
	TEHP	Median/df Range < MDL-22.100 < MDL-10	8.58-1050 < MDL-9.55	315/86300 < MDL-27.7	4.91-1930 < MDL-3	3240-91,000 < MDL-592	982-234,000 < MDL-1690	7.44-7430 < MDL-1050	4.41-139 < MDL-28.7	7.40/76 54.8/100	64.5/97 < MDL-173	97.9/100 29.0-585	10.0/89 < MDL-797
	TCEP	Median/df Range < MDL-22.100 < MDL-10	< MDL-922 < MDL-9.55	12.8-939 < MDL-27.7	< MDL-380 < MDL-3	< MDL-424 < MDL-592	17.9-2930 < MDL-1690	< MDL-1320 < MDL-1050	< MDL-29.8 < MDL-28.7	12.8-322 105/100	< MDL-173 < MDL-156	330/100 164-7530	43.4/100 6.23-2460
	TCIPP	Median/df Range < MDL-22.100 < MDL-10	1.30-229 < MDL-9.55	98.4-1520 < MDL-27.7	2.40-647 < MDL-3	< MDL-28000 < MDL-592	192-15,400 < MDL-1690	< MDL-18500 < MDL-1050	< MDL-139 < MDL-28.7	25.9-1370 745/100	153-6500 1000/100	3070/100 278-66,700	40.5/89 < MDL-2310
	TDCIPP	Median/df Range < MDL-22.100 < MDL-10	< MDL-835 < MDL-9.55	112-6480 < MDL-27.7	< MDL-1100 < MDL-3	47.1-34,300 < MDL-592	78.9/73 < MDL-1690	754/100 < MDL-1050	< MDL/40 < MDL-28.7	81.0/96 203/100	899/100 133-29,200	1750/100 553/100	15.1/72 39.2/100
Aryl-OPEs	TPHP	Median/df Range < MDL-22.100 < MDL-10	< MDL-691 < MDL-9.55	188-13,100 < MDL-27.7	< MDL-361 < MDL-3	103-166,000 < MDL-592	524/100 < MDL-1690	60.5/88 < MDL-1770	16.8/96 < MDL-28.7	37.4-19,500 128/96	14.7-548 30.4/100	285-17,600 17.3/100	9.28-21,800 7.87/100
	TMPP	Median/df Range < MDL-22.100 < MDL-10	3.52-7620 < MDL-9.55	63.0-4660 < MDL-27.7	< MDL-2240 < MDL-3	132/100 < MDL-592	36.8-25,200 < MDL-1690	18.8/100 < MDL-1050	3.26/92 < MDL-28.7	< MDL-125 < MDL-24.5	30.4/100 < MDL-173	56.8-5780 7.3/100	9.28-21,800 7.87/100
	EHDPP	Median/df Range < MDL-22.100 < MDL-10	1.75-116 < MDL-9.55	6.34-145 < MDL-27.7	< MDL-1150 < MDL-3	30.5-1970 < MDL-592	3.69-168 < MDL-1690	2.63-3970 < MDL-1050	2.04/92 < MDL-24.5	46.7/100 15.8-594	113/100 11.6-1200	70.6/100 5.53-530	16.6/100 0.54-294
	CDPP	Median/df Range < MDL-22.100 < MDL-10	0.83-832 < MDL-9.55	13.4-1050 < MDL-27.7	5.47-97 < MDL-3	15.8-192 < MDL-592	15.4-1490 < MDL-1690	3.58-400 < MDL-1050	3.92/92 < MDL-28.7	100/100 5.86-25,700	28.9/100 2.75-137	34.9/100 2.88-6700	6.42/100 1.01-38,900
	IDDP	Median/df Range < MDL-22.100 < MDL-10	0.20-151 < MDL-9.55	< MDL-202 < MDL-27.7	< MDL-866 < MDL-3	< MDL-421 < MDL-592	< MDL-3870 < MDL-1690	< MDL-433 < MDL-1050	< MDL-59.1 < MDL-24.5	5.86-25,700 16.8/100	2.75-137 12.9/100	2.88-6700 46.9/100	1.01-38,900 3.65/100
	BPDP	Median/df Range < MDL-22.100 < MDL-10	3.81/98 < MDL-9.55	10.9/100 < MDL-27.7	4.13/97 < MDL-3	39.0/100 < MDL-592	170/100 < MDL-1690	8.00/83 < MDL-1050	3.86/92 < MDL-28.7	16.8/100 1.77-116	12.9/100 2.19-334	46.9/100 7.46-500	3.65/100 0.43-70.2
	TBPP	Median/df Range < MDL-22.100 < MDL-10	2.12/88 < MDL-9.55	2.49-92.9 < MDL-27.7	0.98/78 < MDL-3	4.67/100 < MDL-592	6.70/100 < MDL-1690	3.38/79 < MDL-1050	1.70/84 < MDL-24.5	13.6/100 2.77-716	13.6/97 < MDL-78.7	37.3/100 0.87-565	0.82/89 < MDL-52.9
	RDP	Median/df Range < MDL-22.100 < MDL-10	< MDL-21.8 < MDL-9.55	< MDL-79.6 < MDL-27.7	< MDL-608 < MDL-3	0.50-24.2 < MDL-592	1.23-117 < MDL-1690	< MDL-54.7 < MDL-1050	< MDL-9.40 < MDL-28.7	< MDL-26 < MDL-5.73	< MDL-29 < MDL-363	< MDL-358 < MDL-40	< MDL-28 22.0/89
	BDP	Median/df Range < MDL-22.100 < MDL-10	< MDL-58.1 < MDL-9.55	< MDL-5160 < MDL-27.7	< MDL-12.8 < MDL-3	< MDL-8.37 < MDL-592	< MDL-9.04 < MDL-1690	< MDL-10.3 < MDL-1050	2.73/76 < MDL-28.7	< MDL-67.7 < MDL-122	3.07/65 < MDL-122	< MDL-95.6 < MDL-5480	< MDL-89 52.6/100
	V6	Median/df Range < MDL-22.100 < MDL-10	< MDL-192 < MDL-9.55	< MDL-101 < MDL-27.7	< MDL-330 < MDL-3	< MDL-60.6 < MDL-592	< MDL-1360 < MDL-1690	< MDL-830 < MDL-1050	0.72/52 < MDL-28.7	< MDL-43 < MDL-26.2	13.0/94 < MDL-668	4.95/87 6.80-1390	< MDL-100 52.6/100
Oligo-OPEs	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100
	ΣAlkyl-OPEs	Median/df Range < MDL-22.100 < MDL-10	< MDL-114 < MDL-9.55	< MDL-63 < MDL-27.7	< MDL-3 < MDL-3	25.3/79 < MDL-592	18.4/97 < MDL-1690	3.45/71 < MDL-1050	< MDL-2.50 < MDL-28.7	< MDL-292 < MDL-57	11.6-319 < MDL/97	< MDL-163 < MDL-39	< MDL-96.6 171/100

^a Sample number.^b Detection frequency (%).

the dominant aryl-OPE, found at a median concentration of 112 ng/g ($< \text{MDL}$ -25,200 ng/g), accounting for 61% of the total aryl-OPE concentrations, which was followed by EHDPP, TMPP, CDPP, IDDP, and BPDP, found at median concentrations of 27.3, 16.5, 15.7, 8.21, and 3.88 ng/g, respectively. It was reported that TPHP was the most widely produced and used aryl-OPE in the US and European countries (Norway, Sweden, Denmark, and Finland) (Van der Veen and de Boer, 2012) and is used as a flame retardant in cellulose acetate, vinyl copolymers in vacuum cleaner parts, computer monitors, and television sets (Green et al., 2008; Saito et al., 2007). Besides, TPHP is an impurity in the commercial mixture of other aryl-OPEs, which can contribute to the increase in TPHP concentrations in indoor dust (Behl et al., 2016; Kademoglou et al., 2017). It should be noted that three novel aryl-OPEs, IDDP, CDPP, and BPDP, were frequently found in house dust samples ($> 90\%$), indicating their widespread global usage. TBPHP, a minor component of commercial BPDP mixture (US EPA, 2008), was detected in only 23% of the house dust samples at a median concentration below the MDL.

3.1.4. Oligo-OPEs

In comparison to monomeric OPEs, oligo-OPEs have lower volatility and higher environmental stability (Van der Veen and de Boer, 2012). There is a general trend that oligo-OPEs may replace monomeric OPEs as flame retardants in the future (European Commission, 2011). For instance, V6 is an alternative to TCIPP and TDCIPP, while RDP and BDP are often used as substitutes of TPHP (Kademoglou et al., 2017). Generally, due to their limited current market share and low mobility (Fang et al., 2013; Liang et al., 2018), oligo-OPEs are rarely detected in the environment. BDP, which is used in furniture, housing for electric and electronic equipment, textiles, and flooring, was found in 73% of the indoor dust samples at a median concentration of 3.45 ng/g ($< \text{MDL}$ -1390 ng/g). The other two oligo-OPEs, V6 (median: 0.75 ng/g) and RDP (median $< \text{MDL}$), were found in only 51% and 47% of the indoor dust samples, respectively. Limited studies, however, have reported high concentrations of oligo-OPEs in house dust from China (Tan et al., 2018), Norway, and the UK (Kademoglou et al., 2017). In addition, an increase in oligo-OPE concentration in river sediments has been reported during the period 2012–2014 in Vietnam (Matsukami et al., 2017).

3.2. Spatial distribution

Statistically significant differences ($p < 0.05$) in ΣOPE concentrations were found in indoor dust samples collected among the 12 countries from 2010 to 2014. The median concentrations of OPEs were the highest in dust samples from South Korea (31,300 ng/g), Japan (29,800 ng/g), and the US (26,500 ng/g). These values were one or two orders of magnitude higher than those in Greece (7140 ng/g), Saudi Arabia (5310 ng/g), Kuwait (4420 ng/g), Romania (4110 ng/g), Vietnam (1190 ng/g), China (1120 ng/g), Colombia (374 ng/g), India (276 ng/g) and Pakistan (138 ng/g). A strong and statistically significant correlation ($r = 0.862$, $p < 0.01$) was observed between median ΣOPE concentrations in house dust and per capita GDP of the 12 countries studied (Fig. S1). Earlier studies have reported higher concentrations of ΣOPEs in indoor dust from more industrialized countries (the US, Canada, Japan, Belgium, Spain, Sweden, Germany, and the Netherlands) than those from less industrialized nations (China, Egypt, Pakistan, Philippines, Saudi Arabia, and Kuwait) (Ali et al., 2017; Kim et al., 2013; Zhou et al., 2017). In general, the concentrations of OPEs in house dust reflect past and current consumption in different countries. The elevated OPE concentrations found in indoor dust from industrialized countries can be attributed to increasing use of these chemicals driven by the ban on PBDEs and requirement of higher flammability standards (He et al., 2018). It has been reported that approximately 60–70% of the global production and consumption of OPEs was in the US and Europe from 1995 to 2008 (Kim et al., 2017). In

contrast, due to less stringent fire safety regulations in developing countries, the use of OPEs in these countries is currently low (Zhou et al., 2017).

3.3. Correlations and profiles

The Spearman correlations among individual OPEs with detection frequencies over 50% in indoor dust are shown in Table S7. All 16 OPEs showed statistically significantly positive correlations ($0.114 < r < 0.748$, $p < 0.05$) with one another in 341 indoor dust samples, indicating that these compounds may have similar sources of origin and have mixed application patterns (Cao et al., 2014). Most of the OPEs are used not only as flame retardants but also as plasticizers in consumer products, resulting in their concurrent occurrence in the indoor environment (Table S6). The strongest correlations were observed between TNBP and TIBP ($r = 0.748$, $p < 0.01$), probably due to their mixed application in household products. In addition, some commercial mixtures of OPEs contain impurities or byproducts (De Boer et al., 2016; Matsukami et al., 2015b). For instance, TCEP was strongly correlated with V6 ($r = 0.652$, $p < 0.01$), which can be attributed to its occurrence as an impurity in commercial V6 mixture (Fang et al., 2013). TPHP showed a moderate to strong correlation with BDP, IDDP, and BPDP ($0.259 < r < 0.602$, $p < 0.01$) because the former is a known impurity in the technical BDP, IDDP, and BPDP mixtures (Behl et al., 2016; Brandsma et al., 2013; Kademoglou et al., 2017).

The contribution of each of the target compounds (based on median concentrations) to ΣOPE concentrations in dust is presented in Fig. 1. $\Sigma\text{Cl-OPEs}$ accounted for 78% of ΣOPE concentrations in dust from Kuwait, followed by Saudi Arabia (73%) $>$ China (65%) $>$ Romania (41%) $>$ the US (37%) $>$ Vietnam (35%) $>$ Colombia (34%) $>$ India (31%) $>$ Greece (30%) $>$ Japan (25%) $>$ South Korea (20%) $>$ Pakistan (9%). $\Sigma\text{Alkyl-OPEs}$ accounted for 77% of ΣOPE concentrations in dust from South Korea, followed by Japan (73%) $>$ the US (58%) $>$ Greece (52%) $>$ Pakistan (45%) $>$ Colombia (43%) $>$ India (38%) $>$ Romania (36%) $>$ Vietnam (33%) $>$ Saudi Arabia (17%) $>$ China (15%) $>$ Kuwait (12%). $\Sigma\text{Aryl-OPEs}$ accounted for 38% of ΣOPE concentrations in dust from Pakistan, followed by India (29%) $>$ Colombia (23%) $>$ Romania (22%) $>$ China (18%) $>$ Greece (17%) $>$ Vietnam (16%) $>$ Kuwait (9%) $>$ Saudi Arabia (8%) $>$ the US (5%) $>$ South Korea (3%) $>$ Japan (2%). $\Sigma\text{Oligo-OPEs}$ accounted for $< 7\%$ of ΣOPE concentrations in all countries except Vietnam (16%), which were consistent with relatively high concentrations of BDP and RDP reported in soil and sediment from Vietnam (Matsukami et al., 2015a; Matsukami et al., 2017).

Cl-OPEs are suspect carcinogens (Van der Veen and de Boer, 2012). TCEP has been listed as a “known carcinogen” by the Californian Environmental Protection Agency since 1992 (Stubbings et al., 2016) and classified as a Category 2 carcinogen by the European Union (European Union, 2009). Higher concentrations of TCEP found in Korea (median: 2490 ng/g), Japan (960 ng/g) and Saudi Arabia (721 ng/g), require further attention with regard to potential impacts on human exposure and health. The profiles of three Cl-OPEs found in indoor dust exhibited a geographical difference among the 12 countries (Fig. 2). High contributions of TCEP to $\Sigma\text{Cl-OPEs}$ were found in dust from most East and South Asian countries, such as China (82%), Pakistan (76%), South Korea (76%), Vietnam (44%), and India (36%); followed by two Middle Eastern countries, Saudi Arabia (28%) and Kuwait (26%); one South American country, Colombia (25%); and Japan (23%), whereas this ratio was low in North America (the US: 6%), and Europe (Romania: 11%, and Greece: 21%). The collective contributions of TCIPP and TDCIPP, two alternatives of TCEP, presented an opposite trend. The composition profiles of OPEs in house dust reflect their consumption patterns globally. The low proportion of TCEP found in dust from the European Union and the US suggests a decline in the consumption of this compound in recent years. TCEP is no longer produced in the

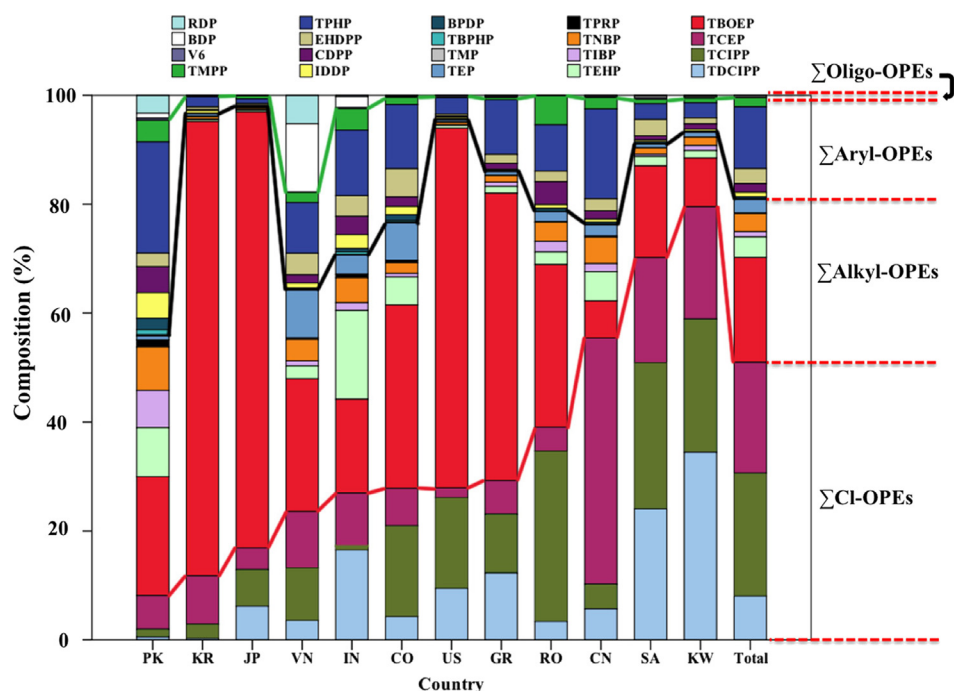


Fig. 1. Composition profiles of organophosphate esters (OPEs) in house dust from 12 countries. Median concentrations were used to calculate the composition profiles.

European Union, and consumption of this compound decreased from 10,500 t in 1992 to 1000 t in 2009 (European Union, 2009). Similarly, production of TCEP in the US dropped sharply from 227–454 t in 2006 to < 11.3 t in 2014 (Van der Veen and de Boer, 2012; US EPA, 2016). The high proportion of TCEP in Asian countries, however, indicates that this compound is still being used, where no regulations currently exist (Zhou et al., 2017). The high proportion of TCEP in house dust from Asian countries warrants further attention.

3.4. Exposure assessment

Previous studies have shown that dust ingestion was the dominant pathway of human exposure to OPEs, while dermal absorption and inhalation were minor contributors to the total daily exposures (Wei et al., 2015). In this study, we calculated the estimated daily intake (EDI) of OPEs via indoor dust ingestion for children and adults (Liao et al., 2012; Wang et al., 2015). The EDI was calculated as shown in Eq. (1).

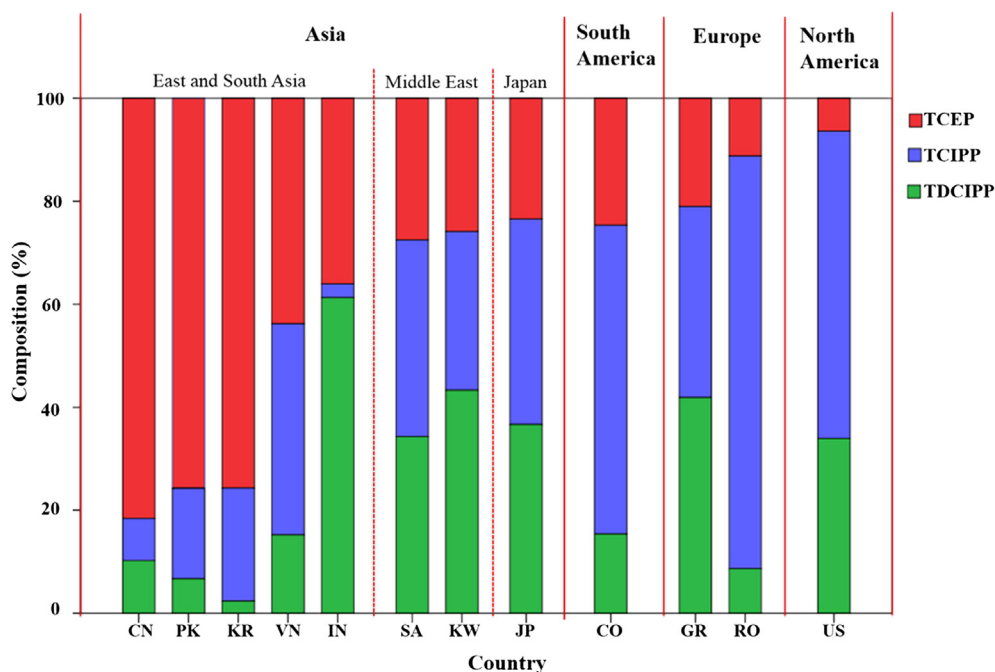


Fig. 2. Composition profiles of TCEP, TCIPP and TDCIPP in house dust from 12 countries. Median concentrations were used to calculate the composition profiles.

Table 3

Estimated daily intakes (EDIs, ng/kg-bw/day) of OPEs (for this with DF > 50%) via indoor dust ingestion for children and adults from 12 countries.

Country	Exposure scenario	TEP	TNBP	TIBP	TBOEP	TEHP	TCEP	TCIPP	TDCIPP	TPHP	TMPP	EHDPP	CDPP	IDDP	BPDP	BDP	V6	ΣOPEs
RfD ^a		13,000	10,000	–	15,000	100,000	7000	10,000	20,000	70,000	13,000	–	–	–	–	–	–	–
<i>Pakistan</i>																		
Children	Average ^b	0.00	0.01	0.01	0.04	0.02	0.01	0.00	0.00	0.03	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.29
	High ^c	0.03	0.03	0.04	0.17	0.05	0.13	0.00	0.06	0.19	0.04	0.04	0.05	0.08	0.02	0.02	0.00	0.92
	Average	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.07
	High	0.01	0.01	0.01	0.04	0.01	0.03	0.00	0.01	0.04	0.01	0.01	0.01	0.02	0.04	0.00	0.00	0.21
<i>India</i>																		
Children	Average	0.01	0.02	0.00	0.06	0.05	0.03	0.00	0.06	0.04	0.01	0.01	0.01	0.01	0.00	0.01	0.00	0.57
	High	0.93	0.07	0.02	0.54	0.34	0.20	0.75	0.33	1.84	0.16	0.11	0.07	0.12	0.05	0.05	0.00	6.64
	Average	0.00	0.00	0.00	0.01	0.01	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.13
	High	0.21	0.02	0.01	0.12	0.08	0.05	0.17	0.08	0.42	0.04	0.03	0.01	0.03	0.12	0.01	0.00	1.53
<i>Colombia</i>																		
Children	Average	0.03	0.01	0.00	0.16	0.02	0.03	0.08	0.02	0.05	0.01	0.02	0.01	0.01	0.01	0.00	0.00	0.70
	High	0.52	0.07	0.01	1.31	0.15	0.35	1.29	0.28	0.50	0.08	0.18	0.05	0.04	0.07	0.03	0.00	4.13
	Average	0.01	0.00	0.00	0.03	0.00	0.01	0.02	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.14
	High	0.10	0.01	0.00	0.26	0.03	0.07	0.26	0.06	0.10	0.02	0.04	0.01	0.01	0.01	0.01	0.00	0.83
<i>China</i>																		
Children	Average	0.03	0.07	0.02	0.09	0.07	0.62	0.06	0.08	0.23	0.03	0.03	0.02	0.01	0.00	0.00	0.00	2.32
	High	0.19	0.42	0.46	0.82	0.44	15.0	0.91	1.92	2.28	0.09	0.11	0.10	0.03	0.02	0.14	0.04	24.6
	Average	0.01	0.02	0.00	0.02	0.02	0.14	0.01	0.02	0.05	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.53
	High	0.04	0.10	0.10	0.19	0.10	3.44	0.21	0.44	0.52	0.02	0.03	0.02	0.01	0.02	0.03	0.01	5.67
<i>Vietnam</i>																		
Children	Average	0.08	0.03	0.01	0.21	0.02	0.09	0.08	0.03	0.08	0.02	0.03	0.01	0.01	0.00	0.11	0.00	2.46
	High	4.45	0.10	0.03	0.35	0.40	1.50	2.86	1.81	11.5	8.34	0.18	14.6	0.08	0.07	1.20	0.04	52.8
	Average	0.02	0.01	0.00	0.05	0.00	0.02	0.02	0.01	0.02	0.00	0.01	0.00	0.00	0.00	0.03	0.00	0.57
	High	1.02	0.02	0.01	0.08	0.09	0.35	0.66	0.42	2.65	1.92	0.04	3.36	0.02	1.07	0.28	0.01	12.1
<i>Romania</i>																		
Children	Average	0.08	0.16	0.09	1.34	0.10	0.20	1.40	0.15	0.38	0.24	0.09	0.19	0.03	0.03	0.00	0.00	7.71
	High	0.34	0.67	0.34	26.4	0.32	1.49	10.3	0.43	33.6	2.44	0.77	14.3	0.18	0.24	0.04	0.08	60.0
	Average	0.02	0.03	0.02	0.27	0.02	0.04	0.28	0.03	0.08	0.05	0.02	0.04	0.01	0.01	0.00	0.00	1.54
	High	0.07	0.13	0.07	5.29	0.06	0.30	2.05	0.09	6.71	0.49	0.15	2.87	0.04	0.01	0.01	0.02	12.0
<i>Kuwait</i>																		
Children	Average	0.04	0.07	0.04	0.41	0.06	0.93	1.11	1.56	0.13	0.04	0.05	0.05	0.02	0.01	0.01	0.01	9.14
	High	0.55	1.96	0.28	10.1	0.69	31.9	39.5	7.22	1.41	1.65	0.48	0.74	0.25	0.07	1.35	0.43	86.5
	Average	0.01	0.02	0.01	0.09	0.01	0.21	0.26	0.36	0.03	0.01	0.01	0.01	0.00	0.00	0.00	0.00	2.10
	High	0.13	0.45	0.07	2.33	0.16	7.33	9.10	1.66	0.32	0.38	0.11	0.17	0.06	0.08	0.31	0.10	19.9
<i>Saudi Arabia</i>																		
Children	Average	0.05	0.09	0.03	1.30	0.13	1.49	2.07	1.86	0.22	0.06	0.23	0.06	0.03	0.03	0.03	0.02	11.0
	High	0.25	0.31	0.05	5.98	0.24	10.4	6.48	17.8	0.77	0.24	1.44	0.23	0.41	0.11	0.91	0.25	31.2
	Average	0.01	0.02	0.01	0.30	0.03	0.34	0.48	0.43	0.05	0.01	0.05	0.01	0.01	0.01	0.01	0.01	2.53
	High	0.06	0.07	0.01	1.38	0.06	2.39	1.49	4.09	0.18	0.06	0.33	0.05	0.10	0.03	0.21	0.06	7.19
<i>Greece</i>																		
Children	Average	0.07	0.12	0.08	5.38	0.13	0.63	1.11	1.25	1.02	0.04	0.17	0.11	0.02	0.02	0.02	0.02	13.4
	High	0.39	0.52	0.43	83.8	0.45	2.04	4.05	7.82	5.53	0.22	1.22	0.40	0.15	0.11	0.57	0.30	103
	Average	0.01	0.02	0.02	1.08	0.03	0.13	0.22	0.25	0.20	0.01	0.03	0.02	0.00	0.00	0.00	0.00	2.68
	High	0.08	0.10	0.09	16.8	0.09	0.41	0.81	1.56	1.11	0.04	0.24	0.08	0.03	0.04	0.11	0.06	20.7
<i>the United States</i>																		
Children	Average	0.14	0.17	0.03	22.9	0.18	0.62	5.76	3.28	1.04	0.03	0.13	0.07	0.09	0.07	0.01	0.12	49.7
	High	0.49	1.57	0.21	70.3	0.86	15.9	55.9	17.8	7.07	0.99	0.68	3.96	0.49	0.52	0.16	2.18	138
	Average	0.03	0.03	0.01	4.58	0.04	0.12	1.15	0.66	0.21	0.01	0.03	0.01	0.02	0.01	0.00	0.02	9.94
	High	0.10	0.31	0.04	14.1	0.17	3.18	11.2	3.56	1.41	0.20	0.14	0.79	0.10	0.02	0.03	0.44	27.6
<i>Japan</i>																		
Children	Average	0.07	0.16	0.02	40.1	0.17	1.99	3.37	3.10	0.41	0.27	0.16	0.09	0.08	0.01	0.01	0.05	61.7
	High	1.17	0.70	0.07	172	0.54	28.8	35.6	146	1.31	1.96	1.76	0.85	0.28	0.05	0.07	18.7	317
	Average	0.02	0.04	0.01	9.24	0.04	0.46	0.78	0.71	0.09	0.06	0.04	0.02	0.02	0.00	0.00	0.01	14.2
	High	0.27	0.16	0.02	39.7	0.12	6.62	8.19	33.7	0.30	0.45	0.41	0.19	0.07	0.02	0.02	4.31	72.9
<i>South Korea</i>																		
Children	Average	0.29	0.27	0.06	48.2	0.19	5.15	1.49	0.16	1.08	0.08	0.27	0.09	0.35	0.01	0.02	0.04	64.8
	High	1.28	1.60	0.25	381	0.70	23.2	7.43	6.87	35.4	0.32	2.23	1.91	14.0	0.16	0.27	0.22	406
	Average	0.07	0.06	0.01	11.1	0.04	1.19	0.34	0.04	0.25	0.02	0.06	0.02	0.08	0.00	0.00	0.01	14.9
	High	0.29	0.37	0.06	87.6	0.16	5.33	1.71	1.58	8.14	0.07	0.51	0.44	3.22	0.14	0.06	0.05	93.3

^a Values are from Li et al. (2018).^b Median concentrations were used for calculating exposure dose.^c 95th percentile (P95) concentrations were used for calculating exposure dose.

$$EDI = \frac{C \times DIR}{BW} \quad (1)$$

where *C* is the median and 95th percentile concentrations of OPEs measured in indoor dust (ng/g), to estimate average and high exposure scenarios, respectively; *DIR* is the dust ingestion rate (g/day), and the average values used in this study were 60 mg/day for children and 30 mg/day for adults; and *BW* is the body weight (kg). The respective average body weights for children and adults in Western countries (the US, Greece, Romania, and Colombia) were 32 and 80 kg, whereas the corresponding values for Asian countries (China, Japan, South Korea, India, Pakistan, Vietnam, Kuwait, and Saudi Arabia) were 29 and 63 kg (Liao et al., 2012).

The EDIs of OPEs via indoor dust ingestion from the 12 countries are summarized in Table 3. The median exposure doses (based on median concentrations) of ΣOPEs in children and adults were in the ranges of 0.29–64.8 and 0.07–14.9 ng/kg bw/day, respectively. Owing to their lower body weights and more frequent hand-to-mouth contact (Hou et al., 2016), children are more highly exposed to OPEs than are adults via dust ingestion. The highest average exposure doses of ΣOPEs through dust ingestion were found in South Korea (adults: 14.9, children: 64.8 ng/kg bw/day), whereas the lowest EDIs were found in Pakistan (adults: 0.07, children: 0.29 ng/kg bw/day). Such regional difference may be related to consumption patterns and the regulation of OPEs among these countries (Hou et al., 2016; Wei et al., 2015).

The highest EDIs found in this study were much higher than those reported from Egypt (adults: 1.14, children: 14.8 ng/kg bw/day) (Abdallah and Covaci, 2014) and Pakistan (adults: 0.78, children: 15.0 ng/kg bw/day) (Ali et al., 2013). However, these EDI values were in the same order of magnitude or lower than those reported in New Zealand (adults: 11.1, children: 259 ng/kg bw/day) (Ali et al., 2013), Belgium (adults: 28.1, children: 548 ng/kg bw/day) (Van de Eede et al., 2011) and Germany (adults: 32.0, children: 160 ng/kg bw/day) (Brommer et al., 2012).

The EDIs of individual OPEs were 3–5 orders of magnitude below the corresponding reference doses (RfDs, Table 3). However, the average and high exposure doses of ΣOPEs in adults were found up to 14.9 and 93.3 ng/kg bw/day, respectively, which were much higher than those reported from drinking water consumption (average: 0.22, high: 1.25 ng/kg bw/day for adults) (Kim and Kannan, 2018), inhalation of particulate matter (average: 0.01–0.59 ng/kg bw/day for adults) (Yang et al., 2014), inhalation of air (high: 34 ng/kg bw/day for adults), dermal exposure from hand wipes (average: 0.32 ng/kg bw/day for adults) (Xu et al., 2016), and daily intake via fish consumption (average: 5.45 ng/kg bw/day for adults) (Kim et al., 2011) and accounted for 2.7–11% of total dietary intake through beverages, dairy products, grains, vegetables, meats, and fresh fruits (average: 539, high: 881 ng/kg bw/day for adults) (Zhang et al., 2016), indicating that indoor dust ingestion is a significant route for human exposure to OPEs.

Despite the fact that we report OPE concentrations and profiles in dust from 12 countries for the first time, this study has some limitations. First, only dust samples from living rooms and bedrooms were collected, whereas dust samples from other indoor microenvironments, such as offices, school buildings, stores, and hospitals, were not included in this study. Second, the sample size for individual countries was small. Nevertheless, we believe that the similar sampling techniques and analytical methods used in this study make direct comparisons of OPE concentrations in dust from various countries more reliable. Information regarding the global distribution of OPEs in the indoor environment is sparse and sporadic, and this study establishes baseline data for comparison with future studies.

4. Conclusions

In summary, this is the first study to report global occurrence and distribution of 20 OPEs in indoor dust from 12 countries. Cl-OPEs were

the predominant OPEs found in indoor dust samples. The variations in ΣOPE concentrations and profiles indicated various consumption patterns and different regulations in regard to the use of OPEs among different countries. Significant correlations among several OPEs in indoor dust samples indicated their similar sources and concurrent applications in consumer products. The median EDI of ΣOPEs via dust ingestion in children and adults was considerably lower than the reference doses, suggesting that the current exposures to OPEs through indoor dust ingestion are not likely to pose risks to human health.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105178>.

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